



## INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 3241-01-WO	<b>FOR FURTHER ACTION</b>		See Form PCT/PEA/416
International application No. PCT/US2004/036671	International filing date ( <i>day/month/year</i> ) 03.11.2004	Priority date ( <i>day/month/year</i> ) 04.11.2003	
International Patent Classification (IPC) or national classification and IPC C08F8/32, C08F10/10, C10L1/22, C10L10/00			
Applicant THE LUBRIZOL CORPORATION et al.			
<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 7 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input checked="" type="checkbox"/> <i>sent to the applicant and to the International Bureau</i> a total of 2 sheets, as follows:</p> <p><input type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).</p> <p><input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.</p> <p>b. <input type="checkbox"/> (<i>sent to the International Bureau only</i>) a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>			
<p>4. This report contains indications relating to the following items:</p> <p><input checked="" type="checkbox"/> Box No. I Basis of the opinion</p> <p><input type="checkbox"/> Box No. II Priority</p> <p><input type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p><input type="checkbox"/> Box No. IV Lack of unity of invention</p> <p><input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p><input type="checkbox"/> Box No. VI Certain documents cited</p> <p><input type="checkbox"/> Box No. VII Certain defects in the international application</p> <p><input type="checkbox"/> Box No. VIII Certain observations on the international application</p>			
Date of submission of the demand  02.09.2005		Date of completion of this report  28.02.2006	
Name and mailing address of the International preliminary examining authority:  European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016		Authorized Officer  Denis, C  Telephone No. +31 70 340-3599 	

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**Box No. I Basis of the report**

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1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
- ☐ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
- ☐ international search (under Rules 12.3 and 23.1(b))
  - ☐ publication of the international application (under Rule 12.4)
  - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements\*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

**Description, Pages**

1-18 as originally filed

**Claims, Numbers**

1-13 received on 02.09.2005 with letter of 02.09.2005

- ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing
3. ☐ The amendments have resulted in the cancellation of:
- ☐ the description, pages
  - ☐ the claims, Nos.
  - ☐ the drawings, sheets/figs
  - ☐ the sequence listing (*specify*):
  - ☐ any table(s) related to sequence listing (*specify*):
4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
- ☐ the description, pages
  - ☐ the claims, Nos.
  - ☐ the drawings, sheets/figs
  - ☐ the sequence listing (*specify*):
  - ☐ any table(s) related to sequence listing (*specify*):

\* If item 4 applies, some or all of these sheets may be marked "superseded."

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**Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

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**1. Statement**

Novelty (N)	Yes: Claims	1-13
	No: Claims	
Inventive step (IS)	Yes: Claims	5,6
	No: Claims	1-4,7-13
Industrial applicability (IA)	Yes: Claims	1-13
	No: Claims	

**2. Citations and explanations (Rule 70.7):**

**see separate sheet**

**Re Item V**

**Reasoned statement with regard to novelty, inventive step or industrial applicability;  
citations and explanations supporting such statement**

Reference is made to the following documents:

D1: DE 199 48 111 A1 (BASF AG) 12 April 2001

D2: EP-A-0 870 819 (ETHYL CORPORATION) 14 October 1998

D3: US-A-5 300 701 (CHERPECK ET AL) 5 April 1994

**1) Novelty (Article 33(2) PCT):**

The subject-matter of claims 1 to 13 of the present application is new in view of documents D1, D2 and D3 in the sense of Article 33(2) PCT:

The document D1 relates to a method for producing Mannich adducts that contain polyisobutylene phenol by: a) alkylating a phenol with highly-reactive polyisobutylene in the presence of an alkylation catalyst; b) reacting the reaction product from a) with formaldehyde and with at least one amine (page 3 lines 1 to 12 and examples Iia to IIf). D1 also relates to Mannich adducts that can be obtained by using this method, to the use of such Mannich adducts as detergent additives in fuel compositions and fuel compositions containing these Mannich adducts (example IV and table 1). The polyisobutylene used in D1 contains more than 80% mole of vinylidene terminal double bond (page 3 lines 14 to 18). D1 does not mention the use of conventional polyisobutylene in the preparation of Mannich adducts.

Document D2 relates to a fuel composition comprising an additive being the Mannich reaction product of 1) an alkylated hydroxyaromatic compound, 2) an aldehyde and 3) an amine. Product 1) is obtained by alkylating hydroxyaromatic compound with polyisobutene (page 2 lines 33 to 48 and page 3 lines 40 to 55). Properties of the additives are assessed in fuel compositions wherein the additive is based on two different types of polyisobutene: conventional polyisobutylene or high reactive polyisobutylene (page 8 lines 12 to 16). Examples of tables 2 show that the use of Mannich reaction products based on high reactive polyisobutylene leads to lower IVD (intake valve deposit) when compared to Mannich reaction products based on conventional polyisobutylene. However, D2 does not disclose any example wherein the fuel additive is derived from high reactive

polyisobutylene **and** conventional polyisobutylene.

Document D3 describes a process for the preparation of polyisobutyl hydroxyaromatic compounds obtained by alkylating phenol with polyisobutylene (column 2 lines 36 to 49). In document D3, the polyisobutylene (PIB) involved in the reaction is a conventional PIB **or** a high reactive PIB (column 4 lines 19 to 43). D3 does not disclose any example wherein the phenol alkylated compound is derived from a mixture comprised of high reactive polyisobutylene **and** conventional polyisobutylene.

The subject-matter of claims 1 to 13 is therefore novel in view of D1, D2 and D3.

## 2) Inventive step (Article 33(3) PCT):

The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1 to 4 and 7 to 13 does not involve an inventive step in the sense of Article 33(3) PCT.

The document D2 is regarded as being the closest prior art to the subject-matter of claims 1 to 13, and discloses a fuel composition comprising an additive being the Mannich reaction product of 1) an polyisobutylene alkylated hydroxyaromatic compound, 2) an aldehyde and 3) an amine. Properties of the additives are assessed in fuel compositions wherein the additive is based on two different types of polyisobutene: conventional polyisobutylene **or** high reactive polyisobutylene. Examples of table 2 show that the use of Mannich reaction products based on high reactive polyisobutylene leads to lower IVD (intake valve deposit) when compared to Mannich reaction products based on conventional polyisobutylene.

### a) Claims 1 to 4 and 7 to 13

The subject-matter of claims 1 to 4 and 7 to 13 of the present application differs from document D2 in that the Mannich additive is a **blend** of a Mannich product derived from a high reactive polyisobutylene and a Mannich product derived from a conventional polyisobutylene.

Examples 4 and 5 of table 1 show that blends of two different Mannich adducts lead to IVD values of 16.5 and 46, i.e. IVD values that correspond to average values of each Mannich adducts when taken individually (IVD=12 for example 2 and IVD=78 for example 3 of table

1).

The problem of the application to be solved may therefore be considered as to provide Mannich reaction products used as additives in fuel compositions being characterised by a IVD value close to that obtained with additives (A) derived from Mannich adducts with high reactive polyisobutylene.

The solution proposed in claims 1 to 4 and 7 to 13 cannot be considered as inventive for the following reasons:

Examples of D2 show explicitly that the use of Mannich reaction products based on high reactive polyisobutylene leads to lower IVD (intake valve deposit) when compared to Mannich reaction products based on conventional polyisobutylene. The skilled man seeking to provide alternative additives for fuel compositions being characterised by a IVD value close to that obtained with additives (A) derived from Mannich adducts with high reactive polyisobutylene, would regard it as normal design to incorporate to such additives (A) a small fraction of similar additives for which the Mannich adduct has been obtained with conventional polyisobutylene. Although D2 does not suggest explicitly to realise such a mixture, this feature (i.e. combining two different Mannich adducts) can be regarded as a normal design for the skilled person in order to solve the problem posed.

Claims 1 to 4 and 7 to 13 are therefore not inventive.

b) Claims 5 and 6

Claims 5 and 6 of the present application differ from D2 in that the polyisobutylene used to prepare the Mannich products is derived from the alkylation of phenol with a **blend** of high reactive polyisobutylene **and** conventional polyisobutylene.

Examples 4 and 5 in table 2 (Mannich product wherein the phenol is alkylated with a blend of two different polyisobutylenes) show improved IVD values when compared to Mannich adducts prepared only with high vinylidene polyisobutylene (examples 1 and 3 respectively of table 2).

The problem to be solved by the present invention may therefore be regarded as to provide alternative Mannich reaction product derived from polyisobutylene for fuel compositions exhibiting low IVD values.

The solution proposed in claims 4 and 5 of the present application can be considered as involving an inventive step (Article 33(3) PCT) for the following reasons:

D2 does not suggest the use of a **blend** of different type of polyisobutylenes (high reactive or conventional) when preparing the Mannich adducts. Moreover, even if the skilled man

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would consider that option obvious, there is no data in D2 demonstrating the surprising effect found in the present application i.e. lower IVD values when using a **blend** of different type of polyisobutylenes (high reactive or conventional) prior to the alkylation or prior to the Mannich reaction when compared to the Mannich adduct obtained using the high reactive polyisobutylene only.

The solution proposed in claims 5 and 6 can therefore be considered as involving an inventive step.

What is claimed is:

1. An additive composition, comprising:  
a Mannich reaction product of  
a) a polyisobutylene alkylated hydroxyaromatic compound;  
b) an aldehyde; and  
c) an amine containing at least one reactive amino group, wherein the said polyisobutylene is derived from a conventional polyisobutylene and a high vinylidene polyisobutylene.
2. The additive composition of claim 1 wherein the conventional polyisobutylene has a trisubstituted double bond isomer content of 45 mole % or greater.
3. The additive composition of claim 1 wherein the high vinylidene polyisobutylene has a combined alpha- and beta-vinylidene double bond isomer content of 70 mole % or greater.
4. The additive composition of claim 1 wherein the polyisobutylene of the alkylated hydroxyaromatic compound has an alpha- and beta-vinylidene double bond isomer content of 50 to 95 mole % and a trisubstituted double bond isomer content of 4 to 40 mole %.
5. The additive composition of claim 1 wherein the said polyisobutylene is derived by combining the conventional polyisobutylene and the high vinylidene polyisobutylene prior to the alkylation of the hydroxyaromatic compound.
6. The additive composition of claim 1 wherein the said polyisobutylene is derived by combining a hydroxyaromatic compound alkylated with the conventional polyisobutylene and a hydroxyaromatic compound alkylated with the high vinylidene polyisobutylene.



7. The additive composition of claim 1 wherein the said polyisobutylene is derived by combining a Mannich reaction product from a hydroxyaromatic compound alkylated with the conventional polyisobutylene and a Mannich reaction product from a hydroxyaromatic compound alkylated with the high vinylidene polyisobutylene.

8. The additive composition of claim 1 wherein the said polyisobutylene has a number average molecular weight ranging from 500 to 3,000.

9. The additive composition of claim 1 wherein the hydroxyaromatic compound is phenol, the aldehyde is formaldehyde or a reactive equivalent thereof, and the amine is a secondary monoamine, an alkylenediamine, or a mixture thereof.

10. A fuel additive concentrate composition for an internal combustion engine, comprising:

a solvent;

the additive composition of claim 1; and

optionally one or more additional fuel additives.

11. A fuel composition for an internal combustion engine, comprising:

a major amount of a fuel; and

a minor amount of the additive composition of claim 1.

12. A fuel composition for an internal combustion engine, comprising:

a major amount of a fuel; and

a minor amount of the fuel additive concentrate composition of claim 10.

13. A method to reduce deposit formation in a fuel system of an internal combustion engine, comprising:

operating the engine with the fuel composition of claim 11.